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Symmetry of superconductivity in fullerides by repulsive interaction model

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Abstract

A repulsive interaction model for superconductivity (SC) in fullerides is studied using a second-order perturbation theory with respect to Coulomb repulsion. With the purely repulsive interaction, T_g and E_g pairs are the most stable in bcc and fcc lattices, respectively. In addition to anisotropic SC pairs, the s-wave (A_g) pair can also be formed in both lattice models. Multiple Fermi surfaces in fullerides stabilize the A_g pairs. We propose that the cooperation between Coulomb repulsion and electron-phonon interaction is responsible for the high T_c.

Keywords: fulleride; multiband; Coulomb repulsion

1. Introduction

The mechanism of the high T_c is discussed by many researchers. Alkali metal doped fullerides (A_3C_{60}) provide an interesting case for such study. A_3C_{60} shows not only superconductivity with high T_c up to about 40K, but also antiferromagnetism with A=Cs. Namely, Cs_3C_{60} is a Mott insulator at ambient pressure [1], while other A_3C_{60} with A = K, Rb are superconductors at low temperature. The insulating state of Cs_3C_{60} changes to superconducting (SC) state under applied pressure, which is in line with other organic superconductor [2]. In view of nearby presence of the metal-insulator transition, A_3C_{60} is a strongly correlated system. It is known that fullerides take two different cubic structures; A15 (bcc like) and fcc lattices [3]. The conduction bands of A_3C_{60} are composed of three-fold degenerate t_{1u} molecular orbitals of a C_{60} [4]. The SC states with orbital degenerate system are attracting recent interest since the discovery of iron pnictide compounds [5].

It has been discussed that the SC state of A_3C_{60} is driven mainly by the electron-phonon interaction. In the conventional theory, the s-wave state is unfavorable in the presence of strong Coulomb repulsion. Han *et al* have pointed out that the dynamic Jahn-Teller effect is responsible for reducing the strong Coulomb repulsion, and thereby stabilizing the ordinary s-wave state [6]. The fundamental question, however, remains why the T_c is so high, since the effective Coulomb repulsion should remain significant even in the SC state.

The interplay of the Coulomb and electron-phonon interactions is a highly delicate matter in the strong-coupling region with high T_c. As a possible step toward the complete understanding, we study purely repulsive models with the characteristic band structure. We discuss here which symmetry is most stabilized in the presence of degenerate orbitals and three dimensional lattice structures.

2. Formalism

To discuss the difference between bcc and fcc lattices, we consider tight binding models which are derived from the three degenerate molecular orbitals of each C_{60}. The hopping integrals are fitted by first principle calculations for

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each lattice model [4, 7]. In this article, we focus on the on-site Coulomb repulsion as a pairing interaction. Taking the second-order perturbation theory according to Ref. [9], we derive the Kohn-Luttinger (KL) interaction for A_3C_{60} as follows [8-10]:

$$V_{mn,m'n'}(k,k') = \left[\, \widehat{U} + \widehat{U}\,\widehat{\chi}(k+k)\,\widehat{U} \,\right]_{mn,m'n'}, \tag{1}$$

$$\chi_{mn,m'n'}(\mathrm{q}) = -\frac{1}{N}\sum_{p,l,l'} \bar{A}_l^{(m)}(p+q)\,\bar{A}_l^{(m')}(p+q)\,A_{l'}^{(n)}(p)\bar{A}_{l'}^{(n')}(p)\,\frac{f(\varepsilon_l(p+q))-f\left(\varepsilon_{l'}(p)\right)}{\varepsilon_l(p+q)-\varepsilon_{l'}(p)}, \tag{2}$$

where *m, n* denote orbital indices, and $\widehat{U}$ is the Coulomb interaction matrix which includes intra-orbital Coulomb U, inter-orbital Coulomb U', the Hund's coupling J, and the pair hopping J'. Here we neglect J and J' and impose the relation $U' = U$ for simplicity. Then the matrix elements exist only for the components: $\widehat{U}_{mn,mn} = U$ (m=n). The other components are set to zero. The matrix $\widehat{\chi}$ is the momentum-dependent static susceptibility, and $\varepsilon_l(p)$ is the energy with l band given by our tight binding model. $A_l^{(m)}(p)$ is a weight of orbital (m) basis in l band. $f(\varepsilon) = 1/(e^{\beta(\varepsilon-\mu)}+1)$ is the Fermi distribution function with μ is the chemical potential and β the inverse of temperature. N is number of lattice points in the system. In order to investigate the SC instability toward the SC state in A_3C_{60}, we solve the following eigenvalue equations [11, 12]:

$$\lambda\,\Delta_l(k) = -\frac{1}{(2\pi)^3}\sum_{k'l'}\frac{dS_{k'l'}}{v_F(k',l')}V_{ll'}(k,k')\Delta_{l'}(k'), \tag{3}$$

$$V_{ll'}(\mathrm{k},k') = \sum_{mn,mn}\bar{A}_l^{(m)}(k)\,\bar{A}_l^{(n)}(-k)\,V_{mn,m'n'}(k,k')A_{l'}^{(m')}(k')\bar{A}_{l'}^{(n')}(-k')\,. \tag{4}$$

where $\Delta_l(k)$ is the SC gap function with band l, $dS_{k,l}$ denotes a Fermi surface (FS) element of the band l with v_F being the Fermi velocity. In the following, we discuss the largest λ which gives the maximum T_c. Since T_c is much smaller than the characteristic energy of $\widehat{\chi}$ in weak coupling region, we put T=0 in Eq. (2).We evaluate the surface integral by using the tetrahedron method [13], and the eigenvalue equation is solved by the power method. In our calculation, more than 10^4 elements on FS are used. Our approach is accurate only for U much smaller than the bandwidth W.

3. Numerical result

We have derived the maximum λ for each symmetry with bcc and fcc crystal structures. In the following, we consider only the case of three conduction electrons per C_{60}. The lattice constant is set to unity. We fix the band width W to 0.5 eV. Then we obtain the simple scaling $\lambda \propto U^2$, provided only the $O(U^2)$ term in Eq. (2) is active. The values of λ computed for each symmetry are summarized in Table 1.

Table 1. The SC coupling constant λ/U^2 for each irreducible representation. The unit of U is eV, which means λ takes the numerical value in the table with U=1 eV. A_g, E_g and T_g pairs are spin singlet states while T_u pair is a spin triplet state.

Crystal structure	A_g	E_g	T_g	T_u
Bcc lattice	0.2358	0.1349	0.6287	0.2860
Fcc lattice	0.2016	0.2926	0.2552	0.1869

Table 1 shows the T_g symmetry of the pair is the most favoured in the bcc model. The eigenstate has the three-fold degeneracy with components (d_{xy}, d_{yz}, d_{zx}). The stability of the T_g pairing is related to the shape of Fermi surfaces and a nesting vector Q_H. We show in Fig.1(a) the gap function $\Delta_l(k)$ of the d_{xy} state along the FS in the k_z=0 plane in Brillouin zone (BZ). The numerical values shown by color mean the amplitude with normalization over the whole FS. The red and blue parts represent positive and negative $\Delta_l(k)$, respectively. The boundary between red part and blue part becomes a node. It is known in the single band bcc model [12] that the SC state with T_g symmetry is most stabilized. With the d_{xy} symmetry, there is substantial combination of momenta that satisfy $k + k' \cong Q_H$. Hence, the effective interaction $V_{ll'}(\mathrm{k},k')$ becomes most effective for the d_{xy} state. Similar situation occurs to other members d_{yz}, d_{zx} of the T_g symmetry. Note that the gap functions of all bands have the same sign for each quadrant of BZ in Fig. 1. This is because each band has comparable size of FS, and compose almost degenerate bands.

On the other hand, Table 1 shows that the SC state with E_g symmetry is the most stable in the fcc model, but the difference with other symmetry is not large. In contrast with the O_h group, which has $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ states as eigenfunctions, the T_h symmetry have no four-fold symmetry. In our result, $\Delta_l(k)$ corresponding to band 1 is close to

$d_{x^2-y^2}$ state, while $\Delta_2(k)$ is rather different from the E_g state in the O_h group. The node structure of $\Delta_l(k)$ in E_g symmetry is complicated.

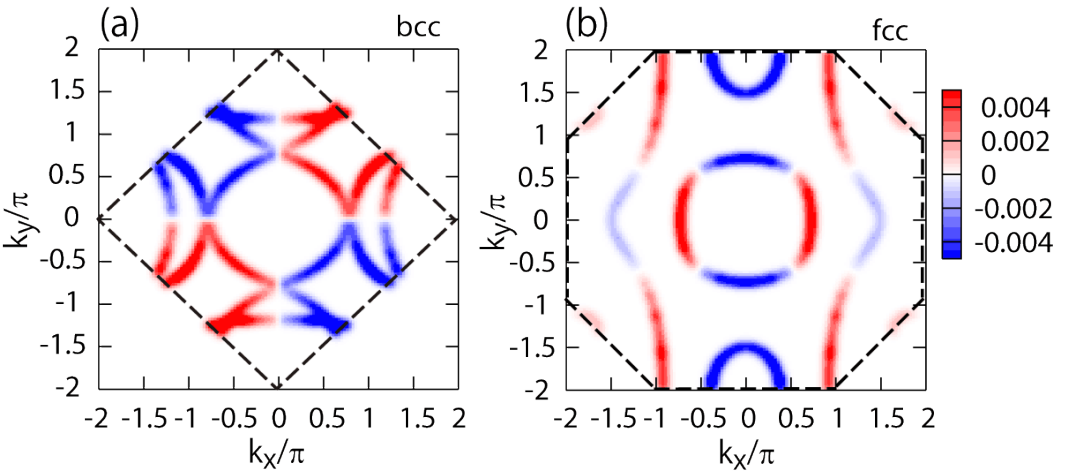

Fig. 1. Illustration of the gap function for the most stable symmetry for each lattice structure, (a) bcc model; (b) fcc model. The gap function is projected onto the plane k_z=0. The dashed lines show the boundary of BZ. The gap function of fcc and bcc models have T_g and E_g, respectively.

We next move to discuss the pairing with the A_g symmetry which is less stable in the present model, but which seems most relevant to actual fullerides. Figure 2 shows the gap function $\Delta_l(k)$ with the A_g symmetry for each structure. Different signs of the gap functions work to cancel the *O(U)* part of the KL interaction. We emphasize such cancellation requires nodes along the FS in the single band model. In our case, bcc model has nodes as in the single band model, while fcc model cancel the *O(U)* without nodes, and hence having less cost in the kinetic energy. In Table 1, the coupling constant λ with A_g symmetry in bcc model is slightly larger than the fcc model.

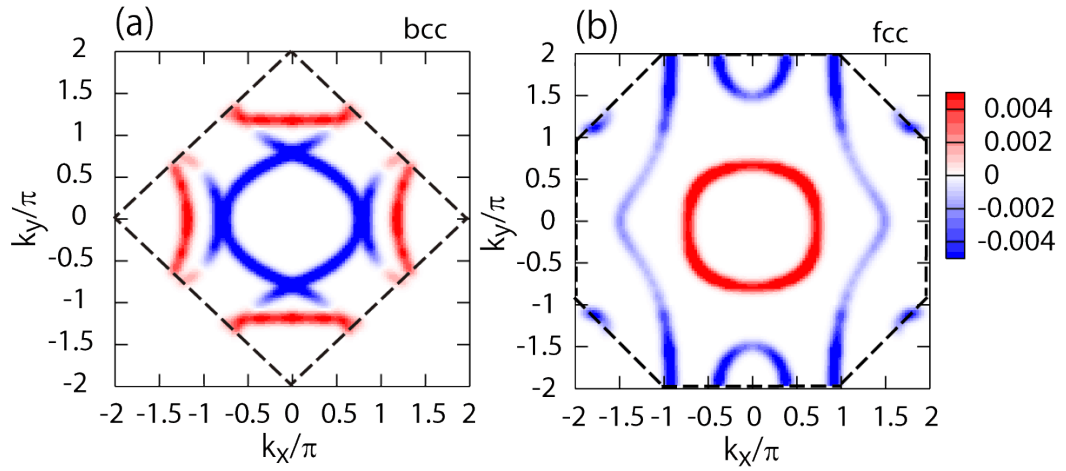

Fig. 2. Illustration of the gap function with A_g symmetry in the k_z=0 plane in each model. (a) In the bcc model, the gap function has nodes keeping the A_g symmetry. (b) In the fcc model, the two sheets of FS have opposite signs of $\Delta_l(k)$.

4. Discussion

We have studied the repulsive interaction model for superconductivity in A_3C_{60}. By using the second order perturbation theory with respect to the Coulomb repulsion, we calculate the SC coupling constant λ for various SC symmetries and for both bcc and fcc lattices. By the nesting property of the FS in the bcc model, the T_g symmetry is the most favorable within the repulsive interaction model. An anisotropic SC state is the most stable also in the fcc model. In the fcc model, various SC symmetries have comparable magnitudes for coupling constant λ. We have further shown that the stable pair with the fully symmetric A_g can also be formed by purely repulsive interaction. We emphasize that the presence of multiple conduction bands is essential for the stability of A_g pair.

Let us now discuss possible relevance of our results to actual superconductivity in fullerenes, especially in Cs_3C_{60} with the highest T_c. The most fundamental question is why the T_c is so high. Since the system is close to the Mott transition, one might naively guess that the SC is caused by a non-phonon mechanism as in cuprate superconductors. However, various experimental evidences exclude the presence of an anisotropic SC state, like d-wave pairing. According to our results, we propose that a following scenario for the SC in Cs_3C_{60}: What is responsible for the high T_c is the cooperation, rather than competition, between the Coulomb repulsion and the Jahn-Teller phonons. The A_g pair will be most favorable for such cooperation. However, it is not clear whether the nodes remain in the presence of Jahn-Teller phonons. If the nodes remain, a gapless SC state is realized. With purely repulsive interaction, the A_g state in the bcc model has nodal lines while there is a full gap in the fcc model. Theoretically, simultaneous account of Jahn-Teller phonons together with Coulomb repulsion requires a new scheme which is not available at present. We have of course recognized that the dynamical mean field theory (DMFT) can powerfully deal with local Coulomb correlation together with Jahn-Teller phonons [6]. The KL interaction is highly non-local and hence is beyond the scope of the DMFT. Various available schemes in the momentum space, on the other hand, are not reliable enough to deal with the case of strong Coulomb repulsion. Namely, SC in fullerenes provides a challenging ground to construct a new theoretical scheme.

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